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Fuel cells: History and updating. A walk along two centuries

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ABSTRACT

This paper reviews the history of fuel cells. Its follows the path from the invention of the fuel cell up to present days. Fuel cell types as well as their advantages, disadvantages and principal applications nowadays are explained. History teaches once again that devices perceived by the public as recent inventions, are actually the product of many years (almost two centuries in this case) of arduous research.

Contents

1. Introduction

Fuel cells are one of the key enabling technologies for future hydrogen economy. For the last 20 years, applications of the fuel cells are mostly replacing internal combustions engines, and providing power in stationary and portable power applications. But the history of the fuel cells is more than last 20 years; actually it has covered almost two centuries. The purpose of this paper is to present the development of the fuel cells across the time, fuel cell types, its present state and some nowadays applications.

This paper is organized in the following sections: in Sections 2 and 3, a review of fuel cell history is done. Section 4 stands today of fuel cells and presents its main applications. Finally, we provide some conclusions, acknowledgements, references and three appendices: (A) Brief biography of the leading inventors, (B) Fuel cells classification and (C) Some current applications, both at the level of prototype as commercial realities.

2. 19th century: the beginnings

There is quite some controversy concerning who discovered the principle of fuel cells. According to the Department of Energy of the United States [1], it was the German chemist Christian Friedrich Schönbein, who in 1838 conducted the first scientific research on the phenomenon of a fuel cell, whose work was published in Philosophical Magazine in the January issue of 1839. By contrast, in reference [2] the author asserts that it was Sir William Robert Grove, who introduced the concept of hydrogen fuel cell. Grove discovered that by immersing two platinum electrodes on one end

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in a solution of sulphuric acid and the other two ends separately sealed in containers of oxygen and hydrogen, a constant current was found to be flowing between the electrodes. Sealed containers contained water together with the respective gases. Grove noted that the water level rose in both tubes as the current flowed. The next step was to realize that combining pairs of electrodes connected in series produced a higher voltage drop, thus creating what he called a gas battery, i.e. the first fuel cell.

The scientific rivalry between Grove and Schönbein lasted many years, as many as their own investigations. A set of letters written by Schönbein to Faraday between 1836 and 1862 can be found in reference [3]. These letters reflect the scientific progress achieved during Schönbein's stay in Bale, which he communicated to his admired colleague Faraday. The editor of these letters, Georg W.A. Kahlbaum, received such letters from Faraday's niece and the Schönbein family. These letters showed the incredulity of the author toward the scientific experiments Grove had underway. It was commented out explicitly that they could not conceive how Grove was able to generate power through oxidation of a positive electrode. This suggests that the original fuel cell comes from Grove and not from Schönbein.

The inverse process to the one occurring in the hydrogen fuel cell, the decomposition of water into hydrogen and oxygen using electricity (water electrolysis), was discovered years before the fuel cell, specifically in 1800 by British scientists Sir Anthony Carlisle and William Nicholson [4]. Both scientists are considered the first ones to produce a chemical reaction using electricity. For their experiment a Volta battery was used. They connected one end of a pair of conducting wires to the electrodes of the battery, while the other end was immersed in a saline solution. The water acted as a conductor and, as a result, hydrogen and oxygen gas were accumulated at the ends of the electrodes.

In 1889, Ludwig Mond and his assistant Car Langer described the process for obtaining nickel (Mond process) with a battery powered gas supplying 6.5 mA/cm^2 (electrode area) to 0.73 V, with perforated platinum electrodes [5].

In 1893, Friedrich Wilhelm Ostwald, considered the founder of the chemistry–physics, experimentally determined the interconnection of various components of a fuel cell: electrodes, electrolyte, oxidizing and reducing agents, anions and cations [6]. At the time, Grove had speculated that the action in his gas battery took place in the contact area between electrode, electrolyte and gas, but there was still quite a lot to be understood. Ostwald, through their pioneering work related to the physical and chemical reactions in fuel cells, solved the puzzle of Grove's gas battery. His research on the chemistry of fuel cells was the basis for further research in this field. In 1896, William W. Jacques [7] developed the first fuel cell with practical applications, and in 1900, Walther Nernst first used zirconium as solid electrolyte.

The Grove's gas battery consisted of 50 monocells with two-dimensional platinum electrodes 31.75 mm wide. The most important observation Grove made, was the need for a significant area of action between gas, electrolyte and electrode. Mond and Langer were the first to refine Grove's cell. The electrodes were porous and three-dimensional shaped, thereby creating the structure of what would be the new generation of fuel cells. With the consolidation of coal as fuel, Mond and Langer realized that it could be used as a source of hydrogen for fuel cells, contrary to Grove's arguments that only pure hydrogen could be used as fuel. Mond, Langer and Ostwald believed that hydrogen could become a common fuel such as coal and that twentieth century would be the beginning of the so-called *Era of Electrochemical Combustion*, where the Rankine's cycle for thermodynamic engines would be replaced by fuel cells (much more efficient and pollution free). But even now, at the twenty-first century, this is not yet realized.

3. 20th century: toward the fuel cell concept

The leading researchers of the late nineteenth and early twentieth century in the field of fuel cells were William W. Jacques and Emil Baur. In 1921, Baur [8] built the first molten carbonate fuel cell (see Appendix B). Jacques was the first one to build high power systems: a 1.5-kW fuel cell with a stack of 100 tubular units, and a fuel cell of 30 kW. During the thirties, Baur experimented with solid oxide electrolytes in high temperature (see Appendix B).

In early 1933, thanks to the developments of Thomas Francis Bacon, one can say that the fuel cell reached its adolescence. Bacon developed the first fuel cell made of hydrogen and oxygen, with practical use. The fuel cell converted air and hydrogen directly into electricity through electrochemical processes. He began his work by investigating alkaline fuel cells (see Appendix B). In 1939, he built a cell with nickel electrodes working at high pressure (200 atm) to prevent the electrolyte to flood the pores of the electrodes. During World War II, Bacon developed a fuel cell to be use in submarines of the Royal Navy. In 1958 he presented to the Britain's National Research Development Corporation an alkaline fuel cell with electrodes of 25.4 mm in diameter. Despite their high cost, Bacon's fuel cells proved to be reliable enough to attract the attention of Pratt & Whitney, the company that acquired the patent of Bacon's work concerning the fuel cell used in Apollo spacecraft. Bacon wanted to use common metals (non-precious metals), a non-corrosive environment (to increase lifetime) and the highest current density with the highest voltage between the electrodes (higher efficiency). In 1959, with the support of the company Marshall Aerospace, he presented a fuel cell of 40 cells of 5 kW and 60% efficiency.

In 1950, a material called teflon (polytetrafluoroethylene or PTFE) began to be available. It was first used in fuel cells with platinum electrodes and acid electrolyte, and with carbon electrodes and alkaline electrolyte. The use of this important material helped developing aqueous electrolyte fuel cells to its current form. In 1955, Thomas Grubb, a chemist who worked for General Electric Company (GE), modified the original design of the fuel cell. For this purpose, he used a membrane made of ion-exchange polystyrene sulphated as an electrolyte. Three years later another GE chemist, Leonard Niedrach, conceived a way of depositing platinum on the membrane (see Appendix B), thus acting as a catalyst for oxidation reactions of hydrogen and oxygen reduction (Fig. 1).

GE developed technology for NASA and McDonnell Aircraft during the Gemini program. In 1959, the team led by Harry Ihrig



Fig. 1. Thomas Grubb (left) and Leonard Niedrach next to their polymer electrolyte membrane fuel cell.



Fig. 2. Fuel cell tractor Allis Chalmers, 1959.

built a fuel cell of 15 kW for a tractor from Allis-Chalmers (Fig. 2). It consisted of a 20 hp tractor with a fuel cell made of 1008 cells of 1 V per cell and potassium hydroxide electrolyte. It used a mixture of gases as fuel, mainly propane and compressed hydrogen, and oxygen acted as an oxidizing agent. This tractor is in the Smithsonian Institute [9]. In the early sixties, attention was focused on the acid electrolyte fuel cell and catalyst platinum in two different ways. One of them employed a polymeric electrolyte, which was simple and reliable. The other form was developed to try to use directly the fuels derived from coal, which was not possible in alkaline fuel cells. This fuel cell type allowed working at high temperatures (150–200 °C). In parallel, scientists G.H.J. Broers and J.A.A. Ketelaar [10,11] abandoned the line of electrolytes oxides, and used molten salts instead. In 1960 they reported having worked for 6 months with a fuel cell using an electrolyte composed of a mixture of lithium carbonate, sodium and/or potassium, impregnated on magnesia sintered porous disk, whose operating temperature reached 650 °C.

In 1961, G.V. Elmore and H.A. Tanner made known a phosphoric acid fuel cell (see Appendix B) in their work entitled *Intermediate Temperature Fuel Cells* [12]. The electrolyte was composed of a mixture of 35% phosphoric acid and 65% of silicon dust stuck to the teflon. They noted that, unlike acid batteries, the electrochemical reduction did not occur during operation of the fuel cell itself. Moreover, this battery could work directly with air instead of pure oxygen. They stated that their pile of phosphoric acid could work for 6 months at 90 mA/cm^2 and 0.25 V without any apparent deterioration. In 1962, J. Weissbart and R. Ruka [13] got working a fuel cell at an operating temperature of 1000 °C and adapted the conducting ceramic oxide impregnated with zirconia of Nerst battery to a solid electrolyte. In 1965, the U.S. Army Engineer Research and Development Laboratory at Fort Belvoir (Virginia), tested various molten carbonate fuel cells manufactured by Texas Instruments.

Fuel cells developed since 1970 have been characterized by the following aspects: suppression of diffusion limitations in the electrodes to obtain a greater area of action (as previously announced by Grove), suppression of the costs of the catalysts, an increased performance and a longer lifetime. There has also been a widespread use of petroleum fuels, using the corresponding fuel-reforming unit. In this way, and taking advantage of the calorific value of these fuels, the efficiency of the phosphoric acid fuel cells reaches 45%, the solid oxide 50% and molten carbonate 60%. A century after the predictions made by Ostwald, the high efficiency, combined with low emissions of harmful gases into the atmosphere, make fuel cells strong candidates for a revolution in the field of electric power generation. The specific power of fuel cell systems has increased considerably over the last 20 years, specifically, two orders of magnitude for the pure hydrogen fuel cells.

In 1990, the Jet Propulsion Laboratory of NASA in conjunction with the University of Southern California developed a battery of methanol (see Appendix B).

4. 21st century: present days

There are now many manufacturers working on fuel cell applications of very different nature [14]. For example, there are many uses of fuel cells with direct application in the automotive sector. The most widespread use can be found in aircrafts, ships, trains, buses, cars, motorcycles, trucks and forklifts. There are also vending machines, vacuum cleaners machines and traffic signals that operate by a fuel cell. On the other hand, there is a growing market for fuel cells for mobile phones, laptops and portable electric devices. At larger scale, there are hospitals, police stations and banks that have a fuel cell system for generating electrical power at their facilities. Water treatment plants and waste dumps are beginning to use fuel cells to carry out the process of converting the methane gas produced for electricity generation. As we can see, the number of different applications of fuel cells is very extensive.

(A) Stationary applications

More than 2500 fuel cell systems have been installed worldwide in hospitals, shelters, centers for elderly care, hotels, offices and schools. In these cases, the fuel cell system is connected to the grid to provide additional electrical power to the plant [15], or as an independent system of the grid to generate electricity in remote or isolated areas [16–19].

Systems of electric power generation based on fuel cells today achieve an efficiency of 40% in the process: using some hydrocarbon as a fuel, and taking as starting point the fuel and as end point electricity generation. Because the fuel cells are working quietly and do not produce environmental pollution, they can be placed near the electricity. Furthermore, in stationary applications, fuel cells are used at high temperature, so the co-generation can increase efficiency up to 85% while reducing energy consumption.

In the field of telecommunications, computers, Internet and telecommunications networks have become essential for humans. This implies the need for a fully reliable electricity supply. Just in this field of application it has been shown that the fuel cell achieved a reliability rate of 99,999% [20]. Thus, fuel cells are starting to compete with batteries for power ranges between 1 and 5 kW in telecommunications systems located at remote places inaccessible to the electrical network. They also use fuel cells to provide power as a primary or support system for telecommunications switching nodes, transmission towers, reception or other electronic devices that can benefit from the DC power supplied by a fuel cell [21].

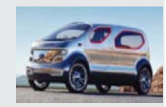
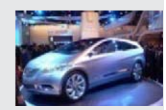
Other applications for stationary fuel cells are in landfills and wastewater plants [22]. The use of fuel cells reduces harmful emissions and allows the generation of power from methane gas (hydrogen-rich fuel used by certain types of fuel cells) that produces the plant. They have also been installed in the breweries of Sierra Nevada (California) and Kirin, Asahi and Sapporo (Japan), to take advantage of the methane produced in the untreated landfills.

(B) Applications for transport

Most vehicle manufacturers are presently using fuel cell vehicles for research, development or testing. Moreover, in 2007, the vehicle manufacturer Honda presented the model FCX Clarity at Los Angeles automobile saloon. This model is available for the consumer since the summer of 2008. This is the first fuel cell vehicle platform-exclusive in the world manufactured in series. Table 1 (see Appendix C) provides a recent summary of models for fuel cell vehicles [23–25].

Regarding buses with fuel cells, in the last 4 years, a number of fuel cell buses have been in operation worldwide. These vehicles are highly efficient even if the hydrogen is produced

Table 1
Recent fuel cell cars.

Manufacturer	Year	Fuel cell	Autonomy/Speed	
Daimler-Chrysler	2008	Hybrid Fuel Cell + Battery	483 km–185 km/h	
Fiat Panda	2007	Nuvera	200 km–130 km/h	
Ford HySeries edge	2007	Ballard	491 km–137 km/h	
GM Provoq	2008	GM	483 km–160 km/h	
Honda FCX Clarity	2007	Honda	570 km–160 km/h	
Hyundai I-Blue	2007	Fuel cell	600 km–165 km/h	
Morgan LIFECar	2008	QinetiQ	402 km–137 km/h	
Peugeot H2Origin	2008	Intelligent Energy	300 km	
Renault Scenic FCV H2	2008	Nissan	240 km–161 km/h	
Mitsubishi SX4-FCV	2008	GM	250 km–150 km/h	
Toyota FCHV-adv	2008	Hybrid Fuel Cell + Battery	830 km–155 km/h	

from fossil fuels and thus reducing CO_2 emissions which would become zero if the hydrogen is produced from renewable sources. In addition, they assist in reducing noise contamination in large cities. Table 2 (see Appendix C) shows recent achievements made with fuel cell buses [26].

Other vehicle using fuel cells is the motorcycle [27]. Despite its small size, motorcycles are a major source of pollution in cities. Motorcycles with two-stroke engines produce a disproportionate amount of emission when compared to its size (almost as much as a diesel truck). Using fuel cells would reduce these emissions.

Fuel cells have been started to be used in electrical conveyor machinery and forklifts. The use of fuel cells in these machines involves a reduction in the cost of logistics, since hardly any maintenance or replacement is needed. Furthermore, due to constant stops and starts, there are many failures and interruptions when using a standard engine. With fuel cells, a continuous supply of power is ensured by eliminating the problems caused by the voltage drop due to the discharge of the batteries.

Another application in the transport sector is the use of fuel cells as auxiliary power units (APUS) in long-haul trucks [28]. Regarding this issue, the Department of Energy of the United States (DOE) estimated about 1.17 billion dollars [29] the annual cost of fuel and engine maintenance for slow motion periods of a truck (parking and rest periods for drivers). During

Table 2
Recent fuel cell buses.

Manufacturer	Year	Fuel cell	Autonomy/Speed	
Volvo	2005	Ballard	563 km–106 km/h	
Mercedes Benz Citaro	2003	Ballard	200 km–80 km/h	
Bavaria	2000	Ballard	300 km–80 km/h	
Neoplan	2000	GmbH	250 km–80 km/h	
Van Hol	2006	UTC	400 km–105 km/h	
Toyota	2001	Toyota	300 km–80 km/h	

this time, the APU is supplying all electricity useful to the driver (heating, air conditioning, computer, TV, radio, refrigerator, microwave, etc.). The Department of Energy states that APUs based on using fuel cells on trucks Mercedes-Benz Class 8 of the whole country could save about 2500 million liters of diesel and between 11 and 80 tonnes of CO_2 per year [30].

Fuel cells are also being developed for mining trains as they do not produce pollution [31]. An international consortium (composed of Vehicle Projects LLC, the U.S. Navy, the National Center for Automotive USA, Inc. Aeroviroment, HERA Hydrogen Storage Systems Inc., Mesofuel Inc., Nuvera Fuel Cells, Jacobs Engineering Group Inc. and the Department of Energy and Natural Resources Canada) is developing since 2003 a locomotive of 109 metric tonnes with a 1.2 MW power plant based on eight modules of the same type of fuel cell PEM 150 kW.

For strategic reasons, the U.S. Navy considers fuel cells a really attractive option for the aircrafts. The use of fuel cells in this case [32] allows reduction of noise emissions from the aircraft, and therefore is not easily detectable with a radar due to the low operating temperature of certain fuel cells, taking advantage of water production also, which can make a significant reduction in the needs of a long-haul aircraft.

The benefits of reduced fuel consumption and increased energy efficiency are the main reasons for considering the use of fuel cells in ships. In general, every litre of fuel consumed in a boat produces 140 times more pollution than a modern car. This has led countries like Iceland to commit them that for 2015 the fishing fleet will use fuel cells to supply auxiliary power. In the second phase, it is also intended to use them as a primary power supply [33]. Table 3 (see Appendix C) provides a summary of the latest applications of fuel cells in transport [34].

(C) Portable applications

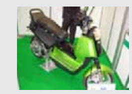
Fuel cells can provide electrical power in places where the grid connection is not available. For example, in a vacation place outdoors (camping area), the use of a fuel cell for electrical power instead of a diesel generator, avoiding harmful emissions, help to preserve the environment and causes no problems noise to other people in the environment [35].

Also, fuel cells are being used as supporting units when power shutdowns occur and in military applications. Fuel cells are much lighter and more durable than batteries, which are particularly important for the soldiers during periods of military maneuvers, and even more in case of war.

(D) Micro power

Fuel cells could change the world of telecommunications, since they can be used in mobile phones or laptops with a lot more battery life [36]. For these applications, and given the characteristics of a fuel cell, the choice is usually a Direct Methanol battery. Companies like Motorola, Toshiba, Samsung, Panasonic, Sanyo and Sony [37] have shown that fuel cells can power telecommunication equipments. For example, they have shown that mobile phones can be run for twice as long as compared to the one that uses a lithium battery with an equivalent size and it needs only 10 min to recharge. As far as laptops are concerned, it has been shown that laptops with fuel cells may be working up to 5 h without refueling. Other applications for micro fuel cells include pagers, video-ReWriters, hearing aids, smoke detectors, security alarms or reviewers counter. In these cases, fuel cells are powered by methanol.

Table 3
Recent fuel cell special vehicles.

Manufacturer	Year	Fuel cell	Autonomy/Speed	
Aerovironment (Global observer)	2005	–	–	
Anuvu (Boat)	2003	PEM	–	
Asia Pacific Fuel cell Technologies (Scooter)	2005	Hybrid Fuel Cell + Battery	80 km–60 km/h	
Astris Energy (Golf car)	2001	Alkaline	10 h–31 km/h	
Besel SA (Wheelchair)	2003	PEM	–	
Deere & Company (Tractor)	2003	Hydrogenic	4 h–50 km/h	

5. Conclusions

Throughout this work we have reviewed the current state of fuel cells as the result of an arduous scientific development of almost two centuries. It is not easy to understand that in times where oil was plentiful, cheap, and when environmental pollution was not an issue of interest (most part of the twentieth century), still the investigators have been working in the field of fuel cells. Very likely, as in other fields of science and technology, military applications and, more recently, space, have sustained the interest

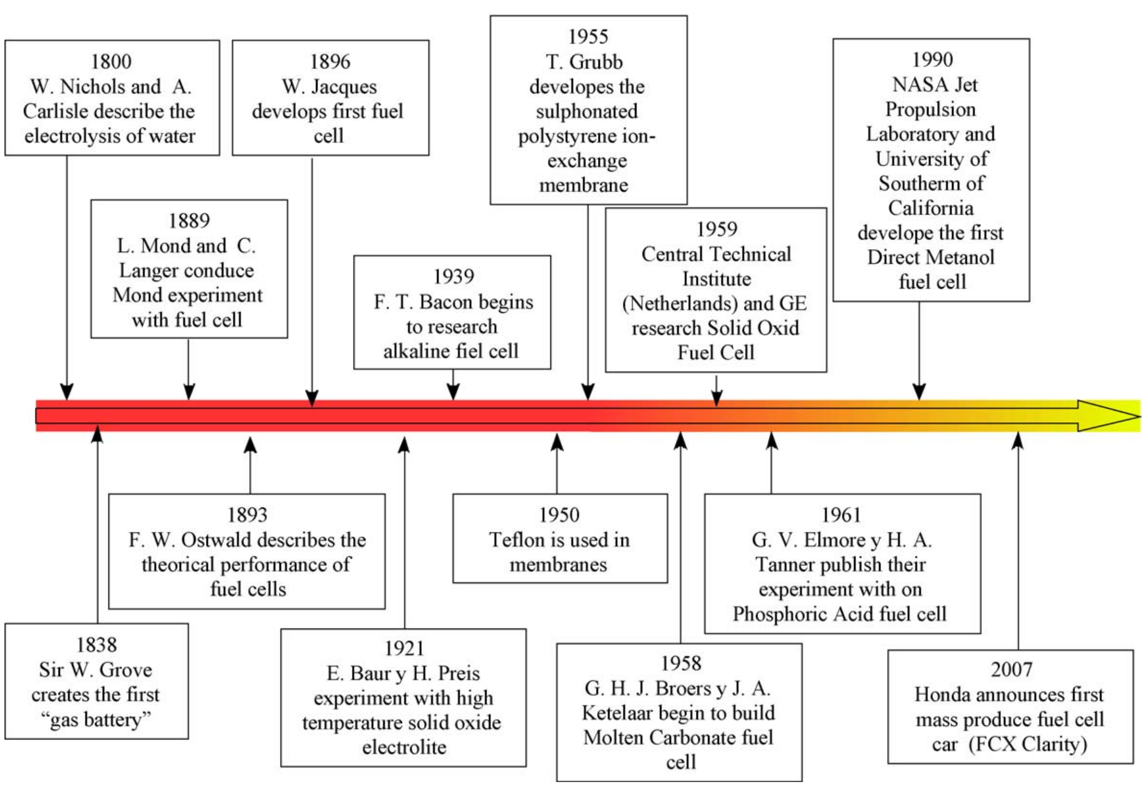


Fig. 3. Historical review of fuel cells.

in fuel cells. Not until today, when oil is scarce, expensive and subject to political vicissitudes, and the Earth is subjected to severe environmental damage, therefore fuel cells are becoming a serious and credible option to substitute other technologies in electricity production.

Although there has been some controversy concerning the authorship in the invention of the fuel cell, several literature sources point to the conclusion that it was Sir William Robert Grove, and not Christian Friendrich Schöbein, the father of the first gas battery or fuel cell. Certainly, the emergence of the fuel cell was not an isolated incidence, but had a strong influence by the earliest discoveries made Sir William Nicholson and Anthony Carlisle concerning the electrolysis of water.

Of the different milestones in the historical development of fuel cells, perhaps the two most important are the theoretical framework developed by Friedrich Wilhelm Ostwald, and their practical implementation by Francis Thomas Bacon. As a summary, in Fig. 3, the reader can find an outline of the major contributions to fuel cell developments over the past two centuries.

Acknowledgement

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Appendix A. The Pioneers



Christian Friedrich Schönbein was born on October 18, 1799 in Metzingen, Swabia (Germany) and died on August 29, 1868, in Sauersberg (Germany). For 13 years he worked as an apprentice to a chemical–pharmaceutical company in Böblingen. As the years went by, he passed the tests to be a chemistry professor at the University of Tübingen. It was in 1828, when he began to work at the University of Basel, where he wrote letters to Michael Faraday and remained until his death. Specifically, the letter he wrote to Faraday Schönbein on April 4, 1840, can be read literally "(. . .) *Our friend Grove wrote me the other day communicating to me some very interesting results of his later researches with which you are no doubt acquainted. What do you think about the fact that the transfer or oxidation of particles in the voltaic arc is definite for a definite current?. Important as such fact is I cannot yet understand it, that is to say, I cannot conceive that by the simple oxidation of the positive electrode the current can be conducted in the same way as by electrolysis, both actions being so very different from each other* (. . .)". To which Faraday replied in his letter of April 24 from Brighton "(. . .) *Your doubts of Grove announcement of a definite transfer matter across air coincide with my own. I cannot deny it but it is so peculiar that it requires the most convincing proofs* (. . .)".



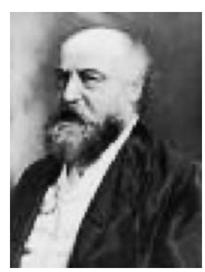
Sir William Robert Grove was born on July 11, 1811 in Swansea (England) and died on August 1, 1896 in London. He was lawyer, judge and physical science expert who anticipated the theory of energy conservation and was a pioneer in the technology of fuel cells. He was the only child of a marriage, whose father was a magistrate and deputy in the chamber of the Lord of Glamorgan. His education was carried out by private tutors before entering the Brasenose College (Oxford) to study humanities, so his scientific interest could be cultivated by the mathematician Baden Powell. In 1835, he joined the Royal Institution and founded the Swansea Literary and Philosophical Society.

Sir Anthony Carlisle was born on February 15, 1768 in Stillington (England) and died on November 2, 1842 in London. He worked as a surgeon. In 1804, he was elected a Fellow of the Royal Society.



William Nicholson was born in 1753 (unknown day and month) and died on May 21, 1815 in Bloomsbury. He was a famous chemist, also known faces of his personality are translator, journalist, publisher, scientist and inventor. After leaving college, he made two trips as a naval officer candidate in the British East India Company. In 1775 he moved to Amsterdam where he worked as a pottery-selling agent. On his return to England, working with Thomas Holcroft developed his talent as a journalist. In 1781, he published his work *An Introduction to Natural Philosophy* and slightly later translation of *the Elements of Newtonian Philosophy* of Voltaire. In 1784, he was secretary of the General Chamber of Manufacturers of Great Britain.

He is considered the inventor of the hydrometer. In 1797, he began to publish the *Journal of Natural Philosophy*, *Chemistry and the Arts*, commonly known as Nicholson's Journal. In 1799, he opened a school in London's Soho district where he taught philosophy and chemistry. In 1788, he wrote First Principles of Chemistry. In 1800, he discovered along with Anthony Carlisle, the process of electrolysis. In 1795, he published the Encyclopaedia Britannica.



Ludwing Mond was born on March 7, 1839 in Kassel (Germany) and died on December 11, 1909 in London. He studied chemistry at the University of Marburg and Heidelberg, but he never completed his studies. He worked in the industry, until in 1862, he moved to Widnes (England) to work in the company John Hutchinson & Co. There he patented a method to obtain sulphide-based products to Leblanc's process (the process for obtaining soda). In 1872, Mond knows Ernest Solvay who develops a better process for obtaining soda (called the Solvay process). Years later, along with John Brunner create Brunner Mond & Company, which was the largest producer of soda in the world for 20 years. However, Mond continued to investigate new chemical processes. He discovered nickel carbonyl (tetracarbonylnickel), hitherto unknown, which could easily break down into pure nickel through the process known as Mond. He founded the Mond Nickel Company, to which he brought the ore from the mines of Canada to Clydach (Wales, Great Britain).



Friedrich Wilhelm Ostwald was born on September 2, 1853 in Riga (Latvia) and died on April 3, 1932 in Grossbothen (Germany). He was a chemist, university professor and philosopher. He obtained German citizenship and was awarded the Nobel Prize in chemistry in 1909. He was educated at the State University of Tartu, where he graduated in 1875 and worked until 1881. From 1881 until 1887, he was professor of the Polytechnic Institute of Riga, where he moved to Leipzig University as a professor of physical chemistry. There he founded the Institute Ostwald (first institute dedicated to the

study of physicochemistry). Ostwald formulated the law that governs the phenomena of dissociation in solutions of electrolysis. In 1900, he discovered a procedure for preparing nitric acid by oxidizing ammonia, facilitating the mass production of fertilizers and explosives in Germany during the First World War. He devised a viscometer that is still used to measure the viscosity of solutions. In the field of philosophy, he emphasized the contribution of energy doctrine that tries to explain most phenomena in terms of their physical energy. His works include *Natural Philosophy* (1902) and *Science of Color* (1923). He won the Nobel Prize for his research on catalysis, the fundamental principles governing chemical equilibria, the reaction rate and chemical equilibrium.

William W. Jacques (1855–1932) was an electrical engineer and chemist. In 1896, he startled the scientific world and general public, according to one scientist of the day, by his broad assertion that he had invented a process of making electricity directly from coal. Jacques constructed a carbon battery in which air was injected into an alkali electrolyte to react with a carbon electrode. It turned out, however, that instead of electrochemical action with efficiency of 82%, he was obtaining thermoelectric action with an efficiency of about 8%.



Walther Nernst was born on June 25, 1864 in Briese (Prussia), and died on November 18, 1941 in Ober-Zibello (Germany). He won the Physical and Chemical Nobel Prize in 1920. He studied at the Universities of Zurich, Berlin, Graz and Wuzburgo. Since 1891 he had worked as a physics professor at the University of Göttingen, where in 1895, he founded the Institute of Chemistry, Physics and Electrochemistry. In 1905, he moved to Humboldt University (Berlin), as the professor and director of the Institute of Chemical Physics. In 1922, he was appointed as the president of the Institute Psychotechnical of Berlin-Charlottenburg, a position he left in 1933 and is dedicated to electroacoustics and astrophysics. Among its well-known contributions are Nerst stress equation (equation which allows the thermodynamic potential between the electrodes of a fuel cell), the heat theorem, the concept of entropy, and the third law of thermodynamics. He also developed a theory to explain and determine osmotic potential between the electrodes of a battery of concentration and formulated the law of distribution of a given area between two phases. He invented the so-called gold ultramicroelectrodes lamp whose filament (consisting of oxides of zirconium and yttrium) is conductor when heated and can reach temperatures above 1000 °C compared to other lamps. They were more effective than the old carbon arc lamps and often used as a source of infrared rays. With his studies of electroacoustics and astrophysics, he invented a microbalance and an electric piano in collaboration with the Bechstein and Siemens companies. Radio amplifiers were used for this purpose. Nerst asteroid was named in his honour, discovered on September 26, 1992 by Freimut Bögmgen and Ludtz D. Schmadel.

Emil Baur (1873–1944) of Switzerland (along with several students at Braunschweig and Zurich) conducted wide-ranging research into different types of fuel cells during the first half of the twentieth century. Baur's work included high temperature devices (using molten silver as an electrolyte) and a unit that used a solid electrolyte of clay and metal oxides.



Francis Thomas Bacon was born on December 21, 1904 at Ramsden Hall, Billericay, Essex (England) and died on May 24, 1992. An engineer, a direct descendant of Sir Nicholas Bacon, was educated at Eton College and Trinity College, Cambridge. He worked as an apprentice to the company C.A. Parsons and Company for the manufacture of turbines. In January 1940, he moved to King's College London laboratory, where he formed a double stack of two units: one unit to generate hydrogen and oxygen (electrolysis process) and the other acting as a fuel cell (producing electricity). The disadvantage of this system was the high operating temperature and pressure and the corrosive nature of the chemical components. In 1946, he moved to the Department of Colloid Science, University of Cambridge. Here, the Bacon team develops wafer samples of porous nickel electrodes for use with large pores on the side in contact with the gas and fine pores on the side in contact with the electrolyte. This helped achieve an electrode–electrolyte interface more stable. During the last years of his life, Bacon was an advisor to the firm Energy Conversion Limited and Johnson Matthey. He was a founding partner of the Fellowship of Engineering and an honorary member of the European Fuel Cell Group.

Appendix B. Fuel cells types

B.1. Molten carbonate fuel cell (MCFC)

The origin of molten carbonate fuel cells lies in other cells such as solid oxide (see section B). During the thirties, E. Baur and H. Preis, experimented with solid oxide electrolytes at high temperature. They found problems with the electrical conductivity and unwanted chemical reactions between the electrolyte and certain gases (including carbon monoxide). Two decades later, scientists H.J. Broers and J.A.A. Ketelaar, seeing the limitations of solid oxides, are focusing their work on molten carbonate salts. In 1960, he announced that he has managed to work for 6 months, a fuel cell that uses as an electrolyte mixture of lithium, sodium and potassium carbonate impregnated in a porous agglomerate disc of magnesium oxide. However, they discovered that the amount of energy produced to the quantity of fuel consumed was less than expected. This was because the junction area of electrode and electrolyte (by the boards), there were losses. In the mid-sixties, the Center for Research and Development Team Mobility U.S. Navy test several molten carbonate fuel cells manufactured by Texas Instruments. These were fuel cells with output power between 100 and 1000 W and were designed to be used in fighting vehicles using an external reformer to extract hydrogen. The Navy wanted to use traditional fuels so that the delivery in the field did not present difficulties.

Molten carbonate fuel cells (Fig. 4) using an electrolyte composed of a mixture of lithium carbonate and potassium. By this electrolyte circulating carbonate ions (CO_3^{2-}) from the cathode to the anode (the reverse of most fuel cells). Fuel cells are working at high temperatures (around 650 °C) and pressures between 1 and 10 atm. Each cell can produce between 0.7 and 1 V. They require carbon dioxide and oxygen as fuels. So far tests have been performed with fuel cells from 10 kW to 2 MW and have been the first to be used in practical applications.

- Advantages:
  - They allow spontaneous internal reforming fuel.
  - Generate a lot of heat.
  - High-speed reactions.

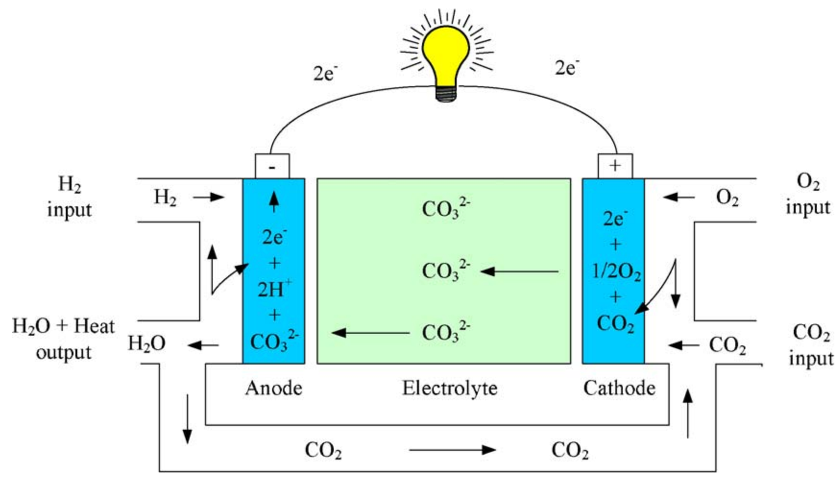


Fig. 4. Molten carbonate fuel cell principles of operation.

  - High efficiency.
  - No need for noble metal catalyst (cost reduction).
- Disadvantages:
  - For further development, they need to be designed using materials resistant to corrosion and dimensionally stable and resistant. The catalyst of nickel oxide cathode can be dissolved in the electrolyte, causing a malfunction. Dimensional instability can cause distortion, changing the active area of the electrodes.
  - They have a high intolerance to sulphur. In particular, the anode does not tolerate more than 1.5 ppm of sulphur particles in the fuel. Otherwise, the fuel cell will suffer a significant deterioration in their functioning.
  - They have a liquid electrolyte, with the corresponding handling problems.
  - They require preheating before starting work.

In view of their ability to work with different types of fuel, MCFC are of great interest. The electrical and operating properties of these fuel cells are sufficient for building economically justified stationary power plants with relatively large power output. The only problem so far is an insufficiently long period of trouble-free operation. The minimum length of time a large (and expensive) power plant should work until replacement is 40000 h (4.5–5 years). In this sense, intense research and engineering effort have made it possible to build individual units that have worked several hundreds and thousands of hours [38]. Yet the road to a guaranteed 5-year period of operation is still long. Many causes lead to a gradual decline in the performance of such power plants, or even premature failure. The three most important reasons associated with fuel cell themselves (rather than with extraneous issues in ancillary equipment or operating errors) are: gradual dissolution of nickel oxide from the oxygen electrode, anode creep and corrosion of metal parts.

B.2. Solid oxide fuel cell (SOFC)

In the late thirties, scientists E. Baur and H. Preis experimented with solid oxide electrolytes, using materials such as zirconium, lanthanum or yttrium.

The SOFCs (Fig. 5) employ an electrolyte that conducts oxide ions (O^{2-}) from the cathode to the anode (unlike other fuel cells). The electrolyte is a solid oxide, zirconium normally. For the electrodes metals such as nickel or cobalt are used. Fuel cells are working at very high temperatures (around 1000 °C) and pressure of 1 atm. Each cell can produce about 0.8 or 1 V. This type of fuel cells used in stationary applications or such as auxiliary power systems (APU).

- Advantages:
  - They allow spontaneous internal reforming fuel. Because the oxide ions travel through the electrolyte, fuel cell can be used to oxidize any combustible gas.
  - Generate a lot of heat.
  - Chemical reactions are very fast.
  - They have a high efficiency.

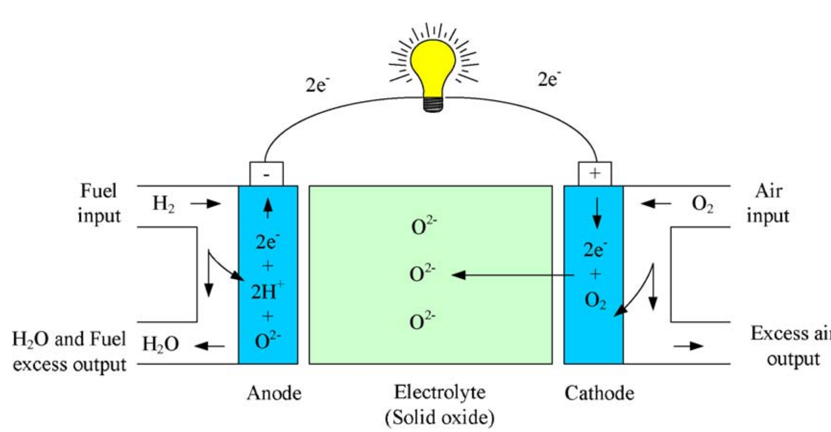


Fig. 5. Solid oxide fuel cell principles of operation.

  - You can work at current densities higher than molten carbonate fuel cells.
  - The electrolyte is solid. Avoids the problems of liquid handling.
  - No need noble metal catalysts.
- Disadvantages:
  - For total market penetration, they need to develop materials that have sufficient conductivity, which remain solid at temperatures of operation, which are chemically compatible with other components of the cell, which are dimensionally stable and have high resistance.
  - They have a moderate intolerance to sulphur (50 ppm).
  - It is not a mature technology.

The major types of fuel for SOFC (as reactants being oxidized) are hydrogen and carbon monoxide. An important difference exists between these fuel cells and other types of fuel cells, in that various natural fuels or products of relatively simple processing of such fuels may also be utilized directly. As we know, the original aim of all work on fuel cells has actually been precisely the direct transformation of the chemical energy of natural fuels to electrical energy. In seeking solutions to this problem of a direct utilization of natural fuel in fuel cells, workers have encountered numerous difficulties that in many cases could practically not be overcome. These difficulties were associated with the very low rates of electrochemical oxidation of these fuels and with the presence of various contaminants that hinder and sometimes block these reactions completely. For this reason, the most realistic way of utilizing these natural fuels in a fuel cell includes their prior chemical conversion to other substances, primarily hydrogen, that are more readily oxidized electrochemically [39]. In addition to conversion, the final processing product must also be carefully free from all contaminants that could hinder the electrochemical reaction.

B.3. Alkaline fuel cell (AFC)

F.T. Bacon was first experimented with alkaline fuel cell using potassium hydroxide electrolyte instead of acids known since the early experiments of Grove. The gas-diffusion electrodes increase the area of reaction between electrode, electrolyte and fuel. Also, Bacon used the compressed hydrogen at high pressure to keep the pores of the electrodes of the aqueous electrolyte. In 1960, NASA and Pratt & Whitney Company established an agreement under the Apollo program to send a spacecraft powered by an alkaline fuel cell. At present, NASA uses this type of fuel cells manufactured by UTC Fuel Cells in the Shuttle program.

Unlike the PEM fuel cells, this type of fuel cells, the electrolyte conducts hydroxide ions (OH^-) from the cathode to the anode (Fig. 6). The electrolyte is composed of a molten alkaline mixture of potassium hydroxide (KOH) which can be mobile or not. In the case of fuel cells with liquid electrolyte, the electrolyte circulates continuously between the electrodes. In fuel cells with fixed electrolyte, the electrolyte is a thin paste adhering to a porous matrix of asbestos. The operating temperature ranges from 65 and 220 °C and pressure of 1 atm. Each cell can deliver about 1.1–1.2 V.

- Advantages:
  - They can work at low temperature.
  - They have a fast start.
  - They have a high efficiency.
  - They use very little amount of catalyst, and thereby lowers costs.
  - They do not have corrosion problems.
  - They have a simple operation.
  - They have low weight and volume.
- Disadvantages:
  - They are extremely intolerant to CO_2 (up to 350 ppm) and show certain intolerance to CO. This limits both the type of oxidant and fuel. Oxididant must be pure oxygen or air free of CO_2. The fuel must be pure hydrogen.
  - Employ a liquid electrolyte, resulting in handling problems.
  - They require an evacuation of the water treatment complex.
  - They have a relatively short lifetime.

The use of AFC in spatial applications is widely known. However, in terrestrial applications, certain difficulties have been encountered. It is natural desire to use air oxygen rather pure oxygen whenever this is feasible. This change is associated, however, with a marked drop in the electrical performance figures relative to those achieved in spacecraft power plants. Also, the complications associated with the presence of CO_2 in the air come into play here. As fuel in AFCs, only pure electrolytic hydrogen is used. It is not possible to use the much cheaper technical hydrogen produced by the reforming of hydrocarbon or other organic compounds. Eliminating CO_2 from hydrogen would be complicated and expensive, a cryogenic storage is not feasible in terrestrial applications and the storage in compressed tanks has weight penalty. All these points and uncertainties as to the potential lifetime, have led to a strongly reduced set of candidate applications for AFC-based power plants. The volume of research and engineering work has shrunk accordingly [40].

B.4. Proton exchange membrane fuel cell (PEMFC)

Polymeric membrane technology was invented by the company GE in the early sixties with the work of T. Grubb and

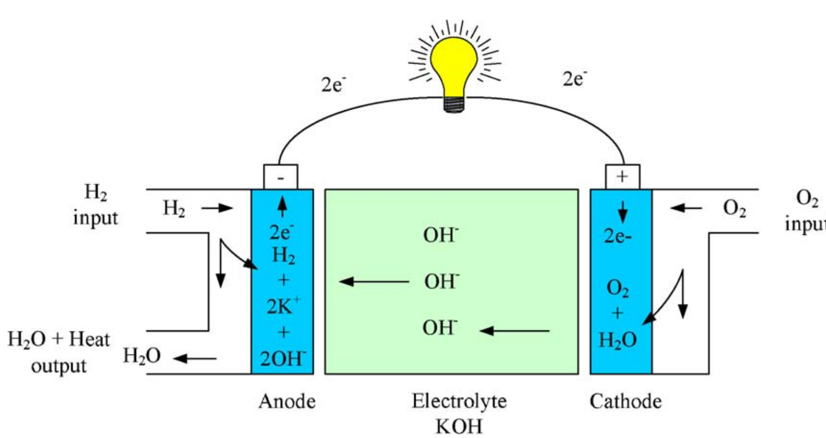


Fig. 6. Alkaline fuel cell principles of operation.

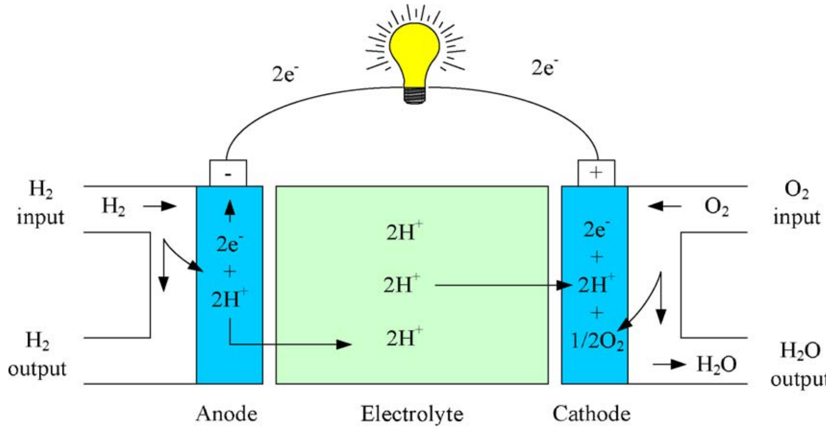


Fig. 7. Proton exchange membrane fuel cell principles of operation.

L. Niedrach. GE announced a first success in the mid-sixties when he developed a small fuel cell for a research program that had with the Electronics Division of the U.S. Navy. This system was fed by hydrogen generated by a mixture of water and lithium hydroxide was stored in bottles. This will facilitate the supply of fuel for activities in isolated areas. The fuel was compact and transportable, but the platinum catalyst raised the price of the system. The operating temperature of these fuel cells is relatively low (around 60–80 °C), so the boot is faster than in high temperature fuel cells. They have high power density and can vary with relative speed their operation point to deliver the required power demand. Among their applications include automotive systems, buildings or portable applications to replace the rechargeable batteries. The maximum power supplied by these batteries ranges from 50 W to 75 kW. Fig. 7 shows the operating principle of a PEM fuel cell type.

- Advantages:
  - Thanks to the separator of anode and cathode is a solid polymer film (planar structure) and that the cell operates at relatively low temperatures, aspects such as handling, assembly or tightness are less complex than in most other types of cells.
  - They use a non-corrosive electrolyte. They remove the need to handle acid or any other corrosive, increasing security.
  - They are tolerant of CO_2; so they can use the atmospheric air.
  - They employ a solid and dry electrolyte so it eliminates the handling of liquids and the problems of resupply.
  - They have high voltage, current and power density.
  - They can work at low pressure (1 or 2 bars), which adds security.
  - They have a good tolerance to the difference of pressure of the reactants.
  - They are compact and robust.
  - They have a simple mechanical design.
  - They use stable building materials.
- Disadvantages:
  - They are very sensitive to impurities of hydrogen, which have developed a number of reforming units to be able to use conventional fuels such fuel cells. PEM fuel cells that directly use methanol as fuel without being reforming are a variant of the direct methanol fuel cell (DMFC).
  - They do not tolerate more than 50 ppm of CO and have a low tolerance to sulphur particles.
  - They need humidification units of reactive gases. If water is used for humidification of gases, the operating temperature of the fuel cell must be less than the boiling water, restricting the potential for cogeneration.
  - They use a catalyst (platinum) and a membrane (solid polymer) very expensive.

Nowadays, PEMFCs are attained a high grade of perfection. They work reliably, exhibit rather good electrical characteristics, and are convenient to handle. A wider use of fuel cells of this type can only be expected when they have conquered two new areas of application: light electric vehicles and portable electronic equipment. For success in this direction, a number of important and rather complex problems must first be solved [41]:

- Longer lifetime for power plants and better stability of the catalysts and membranes.
- Lower cost of production, both for the PEMFC as such for the entire power plant, and the development of catalysts without platinum and of cheaper membranes.
- Higher tolerance of PEMFC for CO impurities in the hydrogen, particularly by building versions of these fuel cells operating at higher temperatures.
- The development of new plants for hydrogen production admitting a wider selection of primary fuels.

B.5. Phosphoric acid fuel cell (PAFC)

Phosphoric acid fuel cells have undergone a slower progress than others due to the low conductivity of the acid. In 1961, G.V. Elmore and H.A. Tanner conducted an experiment with a battery of this type using an electrolyte which was 35% acid and 65% silicon dust stuck on a teflon board. This fuel cell operated with air, not oxygen. In the mid-sixties, the U.S. Navy is studying the possibility of using conventional fuels in PAFC. To do this they used a battery manufactured by Allis-Chalmers and a reformer of Engelhard Industries. Phosphoric acid fuel cells are widely used commercially (installed in many countries in hospitals, hotels, office buildings, schools, water treatment plants). The efficiency of this type of fuel cells reaches 40% in electricity production and 85% in cogeneration. They work about 150–200 °C and pressure of 1 atm. Each cell can produce around 1.1 V. Tolerate the 1.5% impurity of carbon monoxide.

Such fuel cells use an electrolyte that conducts hydrogen ions (H^+) from the anode to the cathode (Fig. 8). The electrolyte is formed, as its name indicates, for a liquid phosphoric acid within a matrix of silicon carbide. There are some fuel cells that use an electrolyte of sulphuric acid.

- Advantages:
  - They tolerate up to 30% CO_2, therefore, these fuel cells may use air directly from the atmosphere.

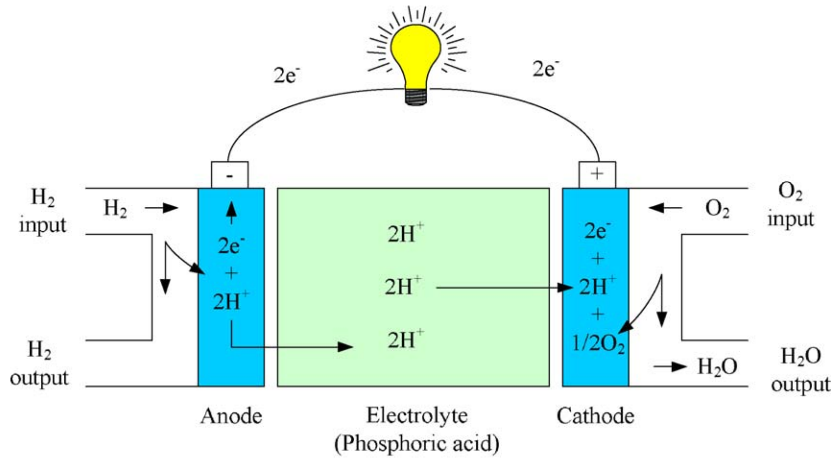


Fig. 8. Phosphoric acid fuel cell principles of operation.

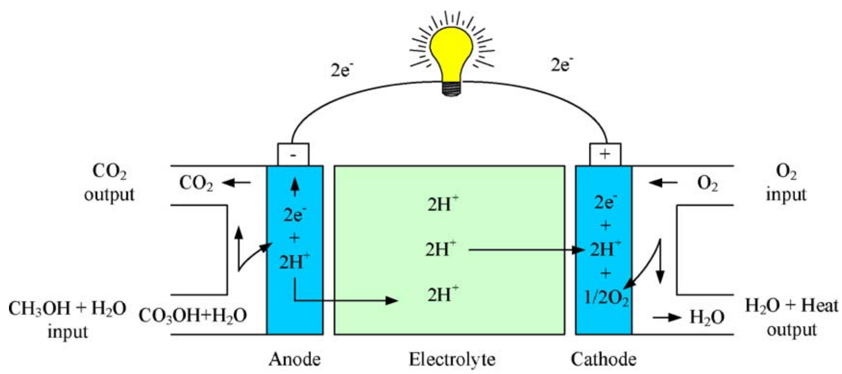


Fig. 9. Direct methanol fuel cell principles of operation.

- While working at medium temperature, they can use the waste heat for cogeneration.
- They use an electrolyte with stable characteristics, low volatility even for temperatures above 200 °C.

• Disadvantages:
- They have a maximum tolerance of 2% CO.
- They utilize liquid electrolyte, witch is corrosive to average temperatures, which involves handling and safety problems.
- They allow the entry of water that can dilute the acid electrolyte.
- They are big and heavy.
- They cannot autoreform fuel.
- They need to reach a certain temperature before starting to work, i.e., they have an operating temperature.

Toward the end of the 1990s, interest en PAFCs gradually waned, despite the success that had been achieved, the relatively large number of intermediate power plants and the installation of several megawatt-sized power plants. On the one hand, this had a strictly economic basis: the high cost of such plants. On the other hand, there were strictly technical problems, that is, insufficient operating reliability in the long term.

During PAFC research and development, technical solutions are being found which are adopted successfully in the development of other fuel cell types. This is true in particular for the use of platinum catalysis, not in a pure form but as deposits on a carbon support, leading to a considerable drop in the amount of platinum needed to manufacture fuel cells [42].

B.6. Direct methanol fuel cell (DMFC)

In 1990, Jet Propulsion Laboratory of NASA in conjunction with the University of Southern California, developed a direct methanol fuel cell. Such fuel cells are replacing traditional batteries in some applications. It is expected to gain space in the market because they have a higher lifetime compared to the lithium ion battery and can be recharged by simply changing the cartridge of fuel. These types of fuel cells are being developed by Samsung (Korea), Toshiba, Hitachi, NEC and Sanyo (Japan). Like PEMFC, these fuel cells use a polymer electrolyte membrane; however, in DMFC anode catalyst extracts hydrogen from liquid methanol, eliminating the need for a fuel reformer. They show efficiencies around 40% and work at temperatures around 130 °C. Applications are employed in small and medium size, to power mobile phones and laptops. Fig. 9 shows the basic functioning of cells MDFC.

• Advantages:
- They use a liquid fuel. The size of the deposits is less and can take advantage of existing infrastructure provision.
- They do not need any reforming process.
- The electrolyte is a proton exchange membrane (similar to the PEM fuel cell type).

• Disadvantages:
- They have low-efficiency with respect to the hydrogen cells.
- They need large amounts of catalyst (noble metal) to the electro-oxidation of methanol at the anode.

In contrast to PEMFCs, and despite the large volume of research performed, DMFC fuel cells are still not in commercial production or in wide practical use. As we have seen, the true performance indicators of these fuel cells when used for different needs are difficult to assess from the experience gathered in tests of individual samples performed under a variety of conditions. Yet even now, one potential area of application can be recognized distinctively: for relatively low-power energy sources in electronic

Table 4
Specific cost for CHP plants with commercial fuel cells.

	PEMFC (BPS)[a]	PAFC (ONSI)[b]	MCFC (MTU)[c]	SOFC (SWPC)[d]
Electrical power	250 kWe	200 kWe	280 kWe	100 kWe
Efficiency	34%	38%	48%	47%
Specific cost, €/kWe	~10000	~5000	~8000	~20000

[a] BPS: Ballard Power System, e.g., BEWAG Berlin 250 kWe PEMFC, natural gas, co-generation.
[b] ONSI: 200 kWe PAFC, natural gas, co-generation.
[c] MTU: Hot module, e.g. Stardwerke Bielefeld, RWE, 250 kWe MCFC, natural gas, co-generation.
[d] SWPC: Siemens WestingHouse Power Corporation, 100 kWe SOFC, co-generation.

equipment such as notebooks, cameras and videocameras, DVV players, and some medical devices. So far, another potential field of application of DMFC as power sources for electric vehicles is too remote. Work is needed to achieve the futures that follow [43]:

- Longer lifetime: the lifetime of DMFC is reduced because of crossover of ruthenium ions and the ageing of methanol adsorption products on platinum.
- Greater efficiency: the efficiency of DMFC is lowered by the methanol crossover. This effect leads to unproductive methanol consumption and to a marked decrease in working voltage caused by the action of methanol on the potential of the oxygen electrode.

Table 4 shows different types of commercial fuel-cell systems of 100–300 kW electrical power, their efficiency as well as specific costs of conventional CHP (Co-generation Heat Power) plants [44].[1]

In addition to the six types of fuel cells viewed, other subtypes are emerging as a result of research and development in some of these fuel cells.

For example, the *Direct Formic Acid Fuel Cell* (DFAFC) which is a subcategory of the PEM fuel cell type. The formic acid (fuel) feds directly to the fuel cell without the need of reforming. Among its applications highlights portable electronic devices such as telephones or portable computers. Such fuel cells have two important advantages with respect to direct methanol fuel cells. On the one hand, storage of formic acid is much simpler and safer than the hydrogen. For another, being liquid at room temperature need not be subjected to high pressures or low temperatures. The chemical reaction that takes place at the anode is the oxidation of formic acid into carbon dioxide and water. At the cathode, the hydrogen ions across the membrane are combined with oxygen to form water.

Direct Ethanol Fuel Cells (DEFC) is a subcategory of PEM fuel cells type, where the ethanol feeds directly to the battery without the need of reforming. Ethanol as fuel is an attractive alternative because it is less toxic and easier to supply the methanol. It is also richer in hydrogen and has a higher energy density (8 kW/kg) than methanol (6.1 kW/kg). It can be obtained in large quantities from biomass by fermentation of renewable resources like sugar cane, wheat or corn. DEFC do not need noble metals as catalysts; moreover, have been achieved power densities of about 140 mW/cm^2 using iron, nickel or cobalt.

Protons Ceramic Fuel Cell (PCFC) uses as electrolyte a ceramic material that shows high proton conductivity at high temperatures. Such fuel cells possess the same kinetic and thermal advantages that the high temperature fuel cells (high-efficiency CHP); at the same time have the ability to oxidize fossil fuels directly without the need of reforming. The gaseous molecules of hydrocarbons (fossil fuels) are absorbed on the surface of the anode in the presence of water vapour, releasing the hydrogen atoms to travel through the electrolyte.

Finally, *Direct Borohydride Fuell Cells* (DBFC), which are considered a subcategory of alkaline fuel cells, use solution sodium borohydride as fuel. The advantage of sodium borohydride from hydrogen in an alkaline fuel cell type is that the use of a highly alkaline fuel and amount of borax (sodium borate) protects the fuel cell of potential carbon dioxide poisoning, i.e., make such fuel cells more tolerant to carbon dioxide that exists in the air.

[1] As it has been mentioned above, AFC and DMFC are not in commercial production, so they do not appear in Table 1.

Appendix C. Nowadays applications

See Tables 1–3.

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